

Quantum Wigner molecules in moiré materials

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The few-body problem (with $N \leq 6$ fermionic charge carriers) in isolated moiré quantum dots (MQDs) in transition metal dichalcogenide (TMD) bilayer materials with integer fillings, $\nu \geq 2$, is investigated by employing large-scale full configuration interaction (FCI; also termed exact diagonalization) computations, and by performing a comparative analysis of the ensuing first-order (charge densities) and second-order (conditional probability distributions, CPDs) correlation functions. With parameters representative of bilayer experimental TMD setups, our investigations reveal the determining role of the strong interparticle Coulombic repulsion in bringing about Wigner molecularization, which is associated with many-body physics beyond both that described by the Aufbau principle of natural atoms, as well as by the widely used Hubbard model for strongly interacting condensed-matter systems. In particular, for weak and moderate trilobal crystal-field deformations of the MQDs, the imperative employment of the CPDs brings to light the geometrical polygonal-ring configurations underlying the Wigner molecules (WMs) that remain hidden at the level of a charge-density analysis, apart from the case of $N = 3$ when a *pinned* WM emerges in the charge density due to the coincidence of the C_3 symmetries associated with both the intrinsic geometry of the $N = 3$ WM and the TMD trilobal crystal field of the confining pocket potential. The FCI numerically exact diagonalization results provide critical benchmarks for assessing and guiding the development of future computational methodologies of interacting strongly correlated fermions in isolated MQDs and their superlattices in TMD materials.

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Understanding of the electronic spectral and configurational organization in natural atoms, which played a pivotal role in the early development of quantum mechanics [1,2], continues to inspire discoveries in research targeting the exploration of the nature of a few charged carriers trapped in artificially fabricated, isolated or superlattice-assembled, quantum dots (QDs) [3–5]. Such research aims at utilizing these systems, with high tunability and control, in future quantum information and computational platforms [6–9]. Earlier studies have unveiled formation of quantum Wigner molecules (WMs), originally predicted theoretically [10–33] in two-dimensional (2D) semiconductor QDs, as well as in trapped ultracold atoms, and subsequently observed experimentally in GaAs QDs [34–37], Si/SiGe QDs [38], and carbon nanotubes [39]. Important contributions to this area employed full configuration-interaction (FCI) [17,18,22,24,28,29,33,40–44] calculations, going beyond (1) the central-field Aufbau principle, which underlies the periodic table of natural atoms [45], and (2) the Hubbard modeling of strongly interacting systems [46,47].

Here we broaden the above WM portfolio, by uncovering, with the use of FCI calculations, the ubiquitous formation of WMs in a novel class of two-dimensional (2D) materials which emerged recently as a most promising platform for investigations of strongly correlated electrons. These materials form moiré superlattices (with large, tunable lattice constants)

when 2D crystals (made of semiconductor or semimetal materials) are stacked with a small twist angle or lattice mismatch; for references to the developing literature on twisted Γ -valley homo-bilayer (e.g., MoS_2) or hetero-bilayer (e.g., WSe_2/WS_2 , $\text{MoSe}_2/\text{WSe}_2$) transition metal dichalcogenides (TMDs), and the emergent twistronics field, see [48–52].

We focus here on few-fermion ($N < 7$, electrons or holes) moiré quantum dots (MQDs) [53–55] formed at the upper layer of doped integer-filled bilayer TMDs [56,57]. The potential confining these fermions at the minima of the 2D moiré superlattice can be approximated by the expression [50–52]

$$V(\mathbf{r}) = -2v_0 \sum_{i=1}^3 \cos(\mathbf{G}_i \cdot \mathbf{r} + \phi), \quad (1)$$

where $\mathbf{G}_i = [(4\pi/\sqrt{3}a_M)(\sin(2\pi i/3), \cos(2\pi i/3))]$ are the moiré reciprocal lattice vectors. The material-specific parameters of $V(\mathbf{r})$ are v_0 (which can also be experimentally controlled through voltage biasing), the moiré lattice constant a_M , and the angle ϕ ; for variations of $V(\mathbf{r})$ with ϕ , see Fig. 1; a_M is typically of the order of 10 nm, which is much larger than the lattice constant of the monolayer TMD material (typically a few Å). The parameter ϕ controls the strength of the trilobal crystal-field-type anisotropy in each MQD potential pocket.

For angles $\phi < 40^\circ$ and a large ratio $v_0/E_K \gg 1$, where the moiré kinetic energy $E_K = \hbar^2/(2m^*a_M^2)$ [51], with m^* being the effective mass of the charge carriers (referred to here, indiscriminately, as “holes”), the attractive pockets of $V(\mathbf{r})$ represent a periodic array of isolated MQDs [53–55],

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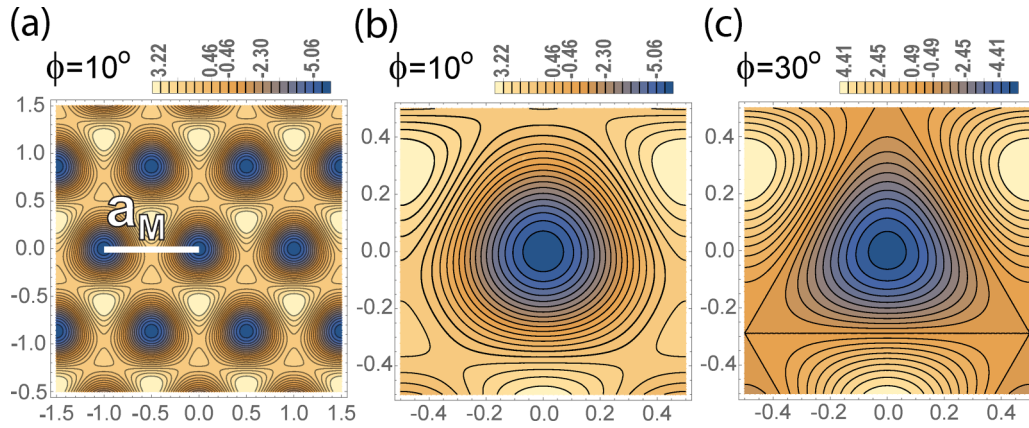


FIG. 1. Plot of the moiré-superlattice potential given by Eq. (1). (a) Broader view of the periodic potential structure for an angle of $\phi = 10^\circ$. (b) Potential of the isolated moiré QD for $\phi = 10^\circ$. (c) Potential of the isolated moiré QD for $\phi = 30^\circ$. Lengths in units of the moiré lattice constant a_M . Potential contours in units of v_0 . Note the change of the length scale in (b) and (c) compared to (a).

where the carriers are tightly bound. In a superlattice with integer fillings, the corresponding strongly interacting few-body problem (with $N \leq 6$ holes) has already been experimentally realized [56,57].

A prerequisite to materialization of gate-controlled tunable MQDs in bilayer TMDs (in isolation or in superlattices) with integer fillings (via doping) in quantum information and simulations is a thorough understanding of the correlated electronic states of the strongly interacting MQD-confined charge carriers. To this aim we concentrate on TMD materials with a large lattice constant, a_M , where the MQDs are evocative of the electrostatically defined ones in 2D semiconductors (SQDs; e.g., in GaAs [22]) with a major difference which is readily grasped by expanding $V(\mathbf{r})$ in Eq. (1) in powers of r , and defining a confining potential, $V_{\text{MQD}}(\mathbf{r})$, for the isolated MQD as follows:

$$V_{\text{MQD}}(\mathbf{r}) \equiv V(\mathbf{r}) + 6v_0 \cos(\phi) \approx m^* \omega_0^2 r^2 / 2 + \mathcal{C} \sin(3\theta) r^3, \quad (2)$$

with $m^* = 0.50m_e$, $m^* \omega_0^2 = 16\pi^2 v_0 \cos(\phi) / a_M^2$, and $\mathcal{C} = 16\pi^3 v_0 \sin(\phi) / (3\sqrt{3}a_M^3)$ [the expansion of $V(\mathbf{r})$ can be restricted to the terms up to r^3]. Because of the anisotropic second term, the $V_{\text{MQD}}(\mathbf{r})$ confinement [Eq. (2)] has a crystal-field, C_3 (trilobal) point-group symmetry and differs from that of the extensively studied single circular 2D QD, which is well approximated solely by the harmonic term [first term in $V_{\text{MQD}}(\mathbf{r})$ above]. Furthermore, consideration of anisotropic confinements in SQDs have treated elliptic-in-shape distortions [using $\omega_{0x} \neq \omega_{0y}$ in place of a single ω_0 and $\mathcal{C} = 0$ in Eq. (2)], under the influence of applied magnetic fields, B [22,31,58,59], lacking a comprehensive study of the trilobal anisotropy in QDs for $B = 0$ (the case of the MQD where diamagnetic effects are negligible due to large values of $\hbar\omega_0$).

Below, we analyze (using representative materials' parameters corresponding to experimentally investigated and theoretically modeled TMD materials [50,51,54,57]) the emergence of quantum moiré WMs (MWMs) in the above trilobal confinement. Specifically, our FCI results will show that a relatively weak $\sin(3\theta)r^3$ term (the case of $\phi < 15^\circ$) can act as a pinning agent for the WM only in the case

of $N = 3$, when the intrinsic WM azimuthal geometry [an empty-center (0,3) polygonal ring] coincides with the C_3 point-group symmetry associated with $\sin(3\theta)$; in this pinned case, the WM is visible in the charge density. In all other instances investigated here, however, there is no pinning [60]; i.e., while the charge densities (CDs) preserve the C_3 symmetry (as quantum-mechanically required), they do not exhibit any signature of Wigner molecularization. Unveiling (seeing) the hidden (unseen) Wigner molecularization necessitates analysis which reaches beyond the information given by the CD distribution in the MQD; that is, it requires analysis of spin-resolved density-density or conditional probability distributions (CPDs; see, e.g., Refs. [12,19,21,22,28,61]; for the definition, see the Supplemental Material (SM) [62]), which reveal the intrinsic (n_1, n_2) (with $n_1 + n_2 = N$, $N = 2-6$) concentric polygonal-ring configurations, familiar from the literature [10,12,14,15,17-19,21-23,25,26,32,33] on circular SQDs.

The Schrödinger equation for the many-body Hamiltonian of the isolated moiré QD, given by

$$H_{\text{MB}} = \sum_{i=1}^N \left\{ \frac{\mathbf{p}_i^2}{2m^*} + V_{\text{MQD}}(\mathbf{r}_i) \right\} + \sum_{i < j}^N \frac{e^2}{\kappa |\mathbf{r}_i - \mathbf{r}_j|}, \quad (3)$$

is solved here using the FCI methodology [17,18,22,33,40,42-44]; for a brief description of this methodology, see the SM [62], including Ref. [63]. The emergence of MWMs is analyzed using both the FCI CDs and CPDs. κ is the effective dielectric constant obtained as the geometric mean of the anisotropic in-plane and perpendicular tensor components of the MQD TMD dielectric environment (most often [50,54,57] hexagonal boron nitride, hBN, when $\kappa \approx 5$). We remark that because the spin and valley degrees of freedom are locked for the holes in TMD materials [50,51,64,65] only the spin needs to be considered in the course of the FCI exact diagonalization of H_{MB} .

For values of $\phi = 10^\circ$ and $\phi = 30^\circ$, $V_{\text{MQD}}(\mathbf{r})$ exhibits, respectively, weak and strong $\sin(3\theta)r^3$ distortions away from the circular confinement ($\phi = 0$); see illustration in Fig. 1. In Figs. 2 and 3, we analyze primarily the $\phi = 10^\circ$ case, with an exception for $\phi = 30^\circ$ in Fig. 2(d). Additional results

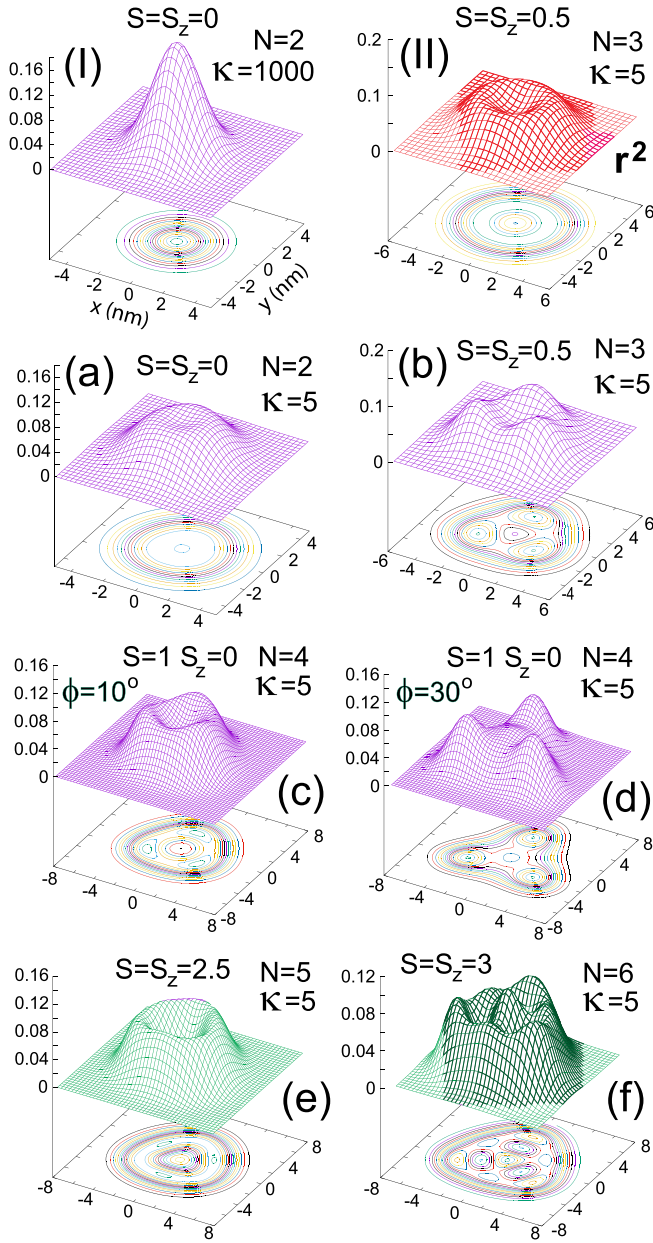


FIG. 2. FCI CDs (normalized to N) for $N = 2$ – 6 holes in the isolated MQD. In all frames: $a_M = 14$ nm, $m^* = 0.5m_e$, $\phi = 10^\circ$, $v_0 = 15$ meV, resulting in $\hbar\omega_0 = 42.59073$ meV [but $\phi = 30^\circ$ in (d)]. $\kappa = 5.0$ ($R_W = 3.57$) except in frame (I), where $\kappa = 1000$ ($R_W = 0.018$) mimics the noninteracting limit. In all cases, the total anisotropic confinement was considered, except in frame (II) (red-colored surface) where the confinement consists solely of the 2D circular harmonic contribution. The green color denotes fully polarized ($S = S_z = N/2$) cases. A vanishing magnetic field, $B = 0$, was considered, except for $N = 3$ and $N = 5$ where a value of $B = 1$ T was used in order to simply lift the ground-state degeneracy. Frames for non-fully-polarized cases describe low-spin ground states. CDs in units of $1/\text{nm}^2$. See the text for a full description.

for $\phi = 30^\circ$, as well as $\phi = 0$ and $\phi = 10^\circ$, are presented in the SM [62]. The Wigner-molecularization propensity uncovered from the FCI results is controlled by the Wigner parameter $R_W = e^2/(\kappa l_0 \hbar\omega_0)$ [10,22], expressing the ratio

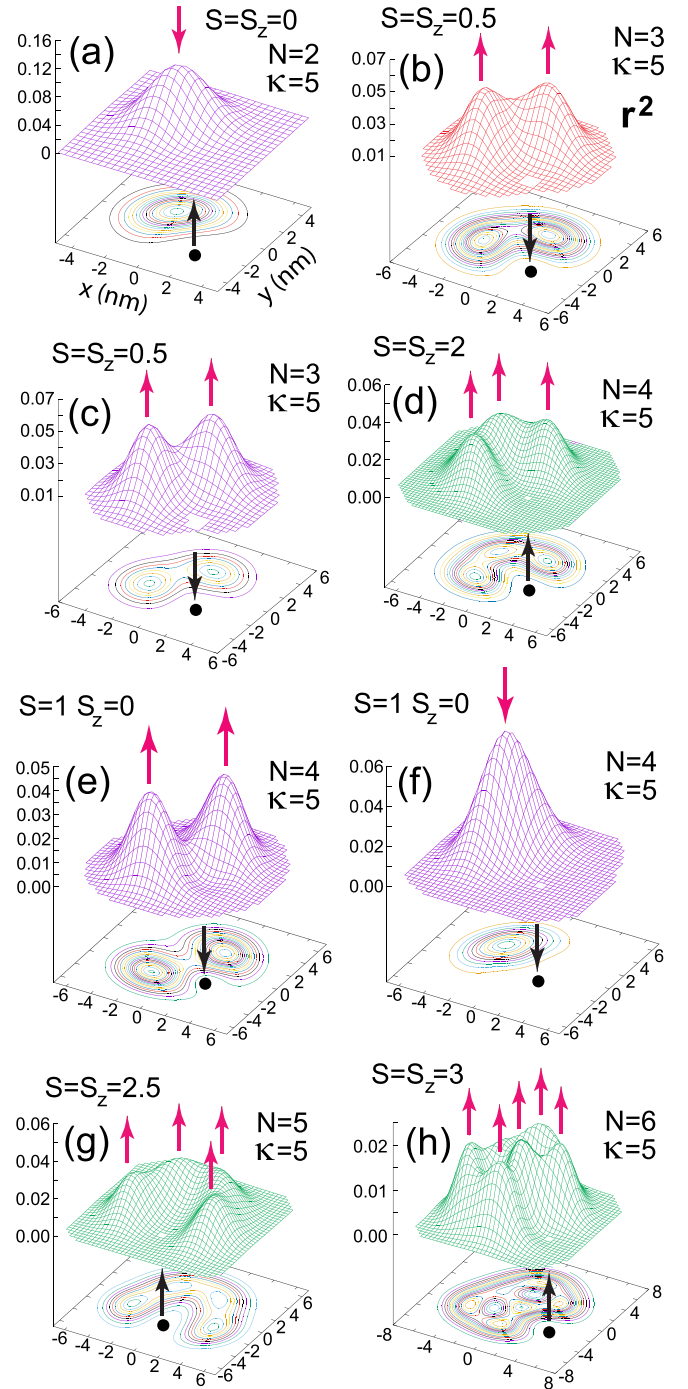


FIG. 3. FCI spin-resolved CPDs (normalized to unity) for $N = 2$ – 6 holes in the isolated MQD. Parameters as denoted (compare Fig. 2). $a_M = 14$ nm, $m^* = 0.5m_e$, $\phi = 10^\circ$, $v_0 = 15$ meV. In all cases, the total anisotropic $V_{\text{MQD}}(\mathbf{r})$ was considered, except in (b) (red-colored surface) where the confinement consists solely of the 2D circular harmonic contribution. The green color denotes again fully polarized ($S = S_z = N/2$) cases, and all the remaining frames describe low-spin ground states. The black solid dot and black arrows denote the position and spin direction of the fixed-point fermions, respectively. The red arrows denote the positions (maxima of the spin surfaces) and spin direction of the remaining $N - 1$ fermions. $B = 0$, except for $N = 3$ and $N = 5$ where a value of $B = 1$ T was used in order to simply lift the ground-state degeneracy. CPDs in units of $1/\text{nm}^2$. See the text for a full description.

between the Coulomb repulsion and the quantum kinetic energy (proportional to the harmonic-oscillator energy gap); $l_0 = [\hbar/(m^*\omega_0)]^{1/2}$ is the oscillator length. A high Wigner molecularization propensity is expected for $R_W > 1$.

Figure 2 displays CDs for $N \leq 6$ in various instances for holes when $\phi = 10^\circ$. For contrast, Fig. 2(d) employs $\phi = 30^\circ$ and in Fig. 2(II) only the circular partial r^2 term has been employed in $V_{\text{MQD}}(\mathbf{r})$. In Fig. 2(I), $\kappa = 1000$ was used to simulate the noninteracting limit, whereas $\kappa = 5$ (strongly interacting regime appropriate for the MQD in an hBN environment) was used in the rest of the frames.

The two frames for $N = 2$ [with $S = S_z = 0$; Fig. 2(I) and Fig. 2(a)] demonstrate the effect of an increasing interparticle Coulomb repulsion (decreasing κ) on the CDs. Indeed, in Fig. 2(I) (with $\kappa = 1000$; noninteracting regime) the CD is associated with a doubly occupied $1s$ orbital. In contrast, for $\kappa = 5$ (MQD/hNB case), the $N = 2$ CD in Fig. 2(a) is spread out over a considerably larger area and it assumes a ringlike shape by developing a depression at the origin. This ringlike shape preserves the C_3 symmetry of the $V_{\text{MQD}}(\mathbf{r})$ by being slightly deformed.

The CD in Fig. 2(a) does not exhibit the anticipated azimuthal geometry of a diatomic molecule, namely two localized particles in an antipodal arrangement. This WM configuration remains hidden in the CD of Fig. 2(a), but it is revealed in the spin-resolved CPD displayed in Fig. 3(a). Indeed, for a spin-up fixed hole, the second hole is found in an antipodal position with an opposite spin, as is consistent for a singlet state ($S = S_z = 0$).

For $N = 3$, the CDs in Fig. 2(II) and Fig. 2(b) exhibit different behaviors. Specifically, when only the isotropic r^2 term is retained in Eq. (2), the CD is an azimuthally uniform ring [Fig. 2(II)]. However, further inclusion of the anisotropic $\sin(3\theta)r^3$ term in $V_{\text{MQD}}(\mathbf{r})$ results in a drastic change; the CD becomes that of three holes localized at the apices of an equilateral triangle [Fig. 2(b)]. One then can talk of a profoundly quantum, “rotating” (0,3) intrinsic WM polygonal-ring structure [66] in the case of Fig. 2(II), which transforms into a *pinned* WM in Fig. 2(b); the intrinsic (0,3) molecular structure is *unseen* in the CD of the isotropic confinement [Fig. 2(II)], but it becomes visible in the spin-resolved CPD in Fig. 3(b). For completeness, we also display [Fig. 3(c)] the spin-resolved CPD for $N = 3$ associated with the total potential, which is redundant, however, since, in this case, the (0,3) molecular configuration is already revealed in the CD [Fig. 2(b)].

With same parameters, the CDs for $N = 4$ (ground state), and for $N = 5$ and $N = 6$ (lowest-in-energy fully spin-polarized states) are displayed in Fig. 2(c), Fig. 2(e), and Fig. 2(f), respectively. These three densities preserve the C_3 symmetry of the external confinement, but they are similar in shape and do not reveal any intrinsic molecular structure. Again, the intrinsic (n_1, n_2) polygonal configurations are revealed through the FCI CPDs. In particular, two spin-resolved CPDs [spin-down at fixed point, look for the two spin-ups, Fig. 3(e), and spin-down at fixed point, look for the other spin-down, Fig. 3(f)], associated with the $N = 4$ ground-state density in Fig. 2(c), are presented. Taking into account the two remaining CPDs (spin-up, look for spin-up and spin-up, look for spin-down, not shown), one concludes that, in this

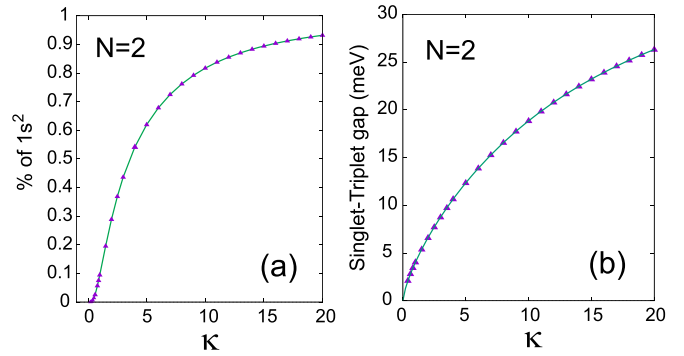


FIG. 4. (a) Weight (%) of the $1s^2$ Slater determinant [an excellent approximation of the non-interacting ground state; see Fig. 2(I)] in the FCI singlet ground state for $N = 2$, as a function of κ . (b) FCI singlet-triplet energy gap for $N = 2$ as a function of κ . Remaining parameters: $a_M = 14$ nm, $v_0 = 15$ meV, $\phi = 10^\circ$, and $m^* = 0.5m_e$. See the text for a full description.

particular case, the intrinsic spin eigenfunction of the rotating WM has the form $(|\uparrow\downarrow\uparrow\downarrow\rangle - |\downarrow\uparrow\downarrow\uparrow\rangle)/\sqrt{2}$ [67], with the localized fermions arranged into a distorted (0,4) polygonal configuration.

In the case of the fully polarized states in Fig. 3(d) ($N = 4$), Fig. 3(g) ($N = 5$), and Fig. 3(g) ($N = 6$), there is only one spin-resolved CPD (spin-up, look for spin-up); in all three cases, this CPD reveals directly the intrinsic structure of a (0,4), (0,5), and (1,5) polygonal WM, respectively. These intrinsic polygonal configurations are necessarily slightly distorted in order to guarantee the C_3 symmetry of the charge densities; the CDs equal the integral of the CPDs over all possible positions of the fixed point [68].

Finally, the CD in Fig. 2(d) for $\phi = 30^\circ$ illustrates that increasing the strength of the crystal-field-like trilobal anisotropy induces a structural isomeric transition [69] of the $N = 4$ MQD from a “rotating” (or gliding) (0,4) WM [Fig. 2(c)] to a pinned (1,3) one.

Using the FCI calculations for $N = 2$, we briefly discuss how the emergence of the Wigner molecularization negates the Aufbau principle that governs the natural atoms. Indeed, the ground state for the noninteracting MQD-helium is approximated by an Aufbau-type single Slater determinant denoted as $1s^2$; see Fig. 2(I). As shown in Fig. 4(a), the weight (%) of the $1s^2$ Slater determinant in the FCI wave function decreases drastically with decreasing κ (increasing correlations), and it vanishes for $\kappa \rightarrow 0$. In the CD plots, this behavior is associated with the developing of a depression at the origin (see Fig. 2(a) and Fig. SF2 in the SM [62]). For $\kappa \rightarrow 0$, singlet and triplet CDs coincide. Furthermore, in accordance, Fig. 4(b) demonstrates that the singlet-triplet energy gap [i.e., the gap Δ_{GLE} between the ground state and the lowest excited one] decreases as well with decreasing κ , and it vanishes for $\kappa \rightarrow 0$. On the contrary, because $N = 2$ is a 2D shell closure, the Aufbau approach would have yielded a value very close to the harmonic-oscillator gap, $\hbar\omega_0$ (i.e., 42.59 meV for the parameters used in Fig. 4).

The voiding of the Aufbau principle with decreasing κ (increasing R_W) can be also seen from the quenching of Δ_{GLE} for $N = 6$, which is also a closed shell in the noninteracting

limit. Indeed, for $\kappa = 3$ and the same other parameters as in Fig. 4, we find values of 1.68 meV, 2.50 meV, and 4.61 meV for the ($S = 0 \rightarrow S = 2$), ($S = 0 \rightarrow S = 1$), and ($S = 0 \rightarrow S = 3$) gaps, respectively. These values are to be contrasted with the noninteracting Δ_{GLE} of $\approx \hbar\omega_0 = 42.59$ meV. This strong quenching of Δ_{GLE} 's conforms with the expected full degeneracy of Wigner crystalline states belonging to a given total-spin multiplicity in the “classical” limit $\kappa \rightarrow 0$ [70].

The FCI second differences, $\Delta_2(N) = E(N+1) + E(N-1) - 2E(N)$, associated with the ground-state energies, relate to the electrochemical potential gap $\Delta\mu$ [71], which is experimentally accessible through measurements of the bilayer-sample capacitance [56]. With respect to the TMD bilayers, the $\Delta_2(N)$ for the isolated MQD relates to capacitance measurements at integer fillings $\nu \geq 1$. We found the FCI values: $\Delta_2(2) = 93.55$ meV, $\Delta_2(3) = 70.73$ meV, $\Delta_2(4) = 78.06$ meV, and $\Delta_2(5) = 60.34$ meV. These values suggest two FCI trends that are in general agreement with the results of Ref. [56]: (i) Due to the Wigner molecularization, the $\Delta_2(N)$ values are substantially smaller than the corresponding Hubbard gap, $U = e^2\sqrt{\pi}/(\kappa l_0) = 269.85$ meV, and (ii) on average, $\Delta_2(N)$ decreases with increasing N [72].

Conclusions. We show that the physics of quantum WMs [10,12,14,15,17–19,21–23,25,26,32,33] underlies at integer fillings $\nu > 1$ that of TMD moiré materials, which as artificial 2D materials are fast developing into a promising experimental platform [56,57], spawning the emergence of the new field of twistrionics [48–52]. Our analysis, using both FCI CDs and CPDs for the single MQD, demonstrates that the anisotropy of the moiré confinement imposes an immediately visible C_3 symmetry on the CDs, while the intrinsic polygonal-ring geometry of the WMs remains hidden and is revealed only using the CPDs. Notable exceptions are the cases of $N = 3$ (at weak) and $N = 4$ (at strong anisotropies)

charge carriers where, due to intrinsic and extrinsic symmetry coincidence, a pinned (0,3) and a pinned (1,3) WM appears in the CDs, respectively; we note here that the discrete structural features of the WMs, having a nested polygonal sliding-ring motif, can be brought to light and accentuated via pinning induced by symmetry-perturbing influences, such as QD shape-distortions (via twist, strain, or gating) or other symmetry-breaking effects [10,29,34]. Formation of the correlated MWMs results in strongly quenched energy gaps compared to those expected from both the Hubbard model and the Aufbau principle. Our FCI results provide essential benchmarks for developing future many-body computational methodologies, and in particular those based on machine learning and artificial intelligence [73]. The quantum WM phases predicted here can be experimentally verified using scanning tunneling microscopy for the CDs [53,74] and scanning probe microscopy for the CPDs [75,76].

Note added. We recently became aware of a preprint by Luo *et al.* [77] in which Wigner molecularization in isolated TMD moiré QDs has been addressed. The results reported in that work differ substantially from our benchmark FCI results. We note in particular the inability of that study, based on neural network methodology, to (1) identify formation of a Wigner molecule for $N = 2$, and (2) recover all the exact charge densities for $N > 3$. In the context of point (2) above, we call attention to pertinent remarks in Ref. [78] about ongoing issues in fermionic neural networks pertaining to Wigner crystallization and the conservation of the symmetries of the many-body Hamiltonian, which is a *sine qua non* for exact many-body solutions.

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